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# **Development of Laminated Fabric Materials**

**American Cyanamid Company**

**prepared for  
Army Natick Laboratories**

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TECHNICAL REPORT

73-21-CE

# DEVELOPMENT OF LAMINATED FABRIC MATERIALS

by

Darwin DeLapp

American Cyanamid Company

Stamford, Connecticut

Contract No. DAAG17-72-C-0080

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UNITED STATES ARMY  
NATICK LABORATORIES  
Natick, Massachusetts 01760



CLOTHING & PERSONAL LIFE SUPPORT EQUIPMENT  
LABORATORY

C&PLSEL-101

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<p>Laboratory and commercial scale experiments produced a three layer laminated fabric (5.2 oz/yd<sup>2</sup>) exhibiting 21.5 psi hydrostatic resistance, and permitting 590 grams per square meter per 24 hours moisture vapor transmission. The functional and processing characteristics of six different porous polymer structures, comprising the inner laminate layer, were explored. The mechanical operation even at full scale proceeded without difficulty. Additional equipment design and development is needed to achieve adequate product quality control and production rate.</p>		

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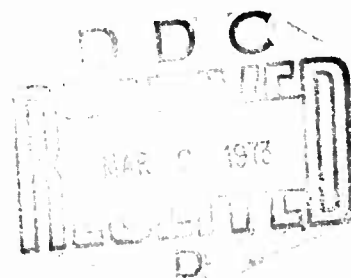
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## TABLE OF CONTENTS

	<u>Page</u>
Abstract	iv
A. Statement of the Problem	1
B. Material Optimization	1
C. Intermediate Scale-Up	4
D. Process Development and Material Production	6
E. Recommendations	12
Summary	16
Appendix A - Procedure for Laboratory Preparation of Laminated Fabric Test Specimens	17
Appendix B - Procedure for Large Scale Preparation of Laminated Sheet	20

## List of Tables

<u>Table</u>	
I Fabric Construction	2
II Laminated Fibrillar Materials	3
III Operating Conditions of WRAPS, Inc. Run 1	5
IV Operating Conditions of WRAPS, Inc. Run 2	9
V Physical Properties of WRAPS, Inc. Run 2	10
VI Operating Conditions of WRAPS, Inc. Run 3	13
VII Pre-Extraction Characteristics of Product of WRAPS, Inc. Run 3	14
VIII Physical Properties of WRAPS, Inc. Run 3	15
Figure 1 Scanning Electron Photomicrographs of Pigmented Feed Polymer Compounded at Werner & Pfleiderer for Extrusion at WRAPS, Inc.	7

#### A. Statement of the Problem

The function of wet weather clothing is to protect personnel from rain and sea spray without reducing their working efficiency or producing discomfort. Known fabric type materials that can be designated "waterproof" in the sense that they prevent the passage of water also excessively impede the transmission of water vapor. Hence, whenever such materials are used for clothing, normal perspiration is trapped close to the wearer's skin and cannot be evaporated. The result is impaired work capacity and heat-induced discomfort.

A requirement exists for materials suitably convertible into various kinds of apparel which will keep a person dry, both by shedding rain and by allowing body moisture to evaporate into the environment.

The American Cyanamid Company had discovered how to make material with potential to meet this requirement.<sup>1</sup> Molding grade poly(methyl methacrylate) pellets, poly(ethylene oxide), isotactic poly(1-butene) pellets and a small quantity of poly(tetrafluoroethylene) in aqueous dispersion form were combined in a ribbon blender at room temperature. The resulting mixture was charged to a twin screw extruder and melt compounded at 200°C. The product was extruded to form a continuous sheet 10 mils thick. The sheet was extracted with methylene chloride to remove substantially all poly(methyl methacrylate) and poly(ethylene oxide). An unsolicited proposal was made to pursue further development and the following research objectives were decided upon:

- a. To optimize the performance characteristics of laminated fabric for wet weather protective apparel by systematic variation of material formulation and fabric construction including pigmentation to an OG106 shade.
- b. To develop techniques to make the best current material 12 to 24 inch width, submitting ten yards to NLABS for evaluation.
- c. To develop a full scale process for the optimized construction, as designated by NLABS, to produce and deliver 100 yards of material of 36-inch width, minimum.

#### B. Material Optimization

1. Six different fibrous porous polymer structures were made by a continuous extrusion-lamination process (See Appendix A). A Brabender laboratory-size extruder equipped with a 4-inch wide ribbon die was used to feed the inner layer to a 3-roll lamination station of Cyanamid's design and construction. Finished fabrics where nylon was used as the surface, encompassed a systematic variation in thickness,

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1. U. S. Patent 3,675,391.



yielding cloth weights of 3.4 to 7 oz/yd<sup>2</sup>, except where poly(tetrafluoroethylene) alone comprised the inner layers. This material could not be processed to the lighter constructions. The composition and appearance of the material is shown in Table I.

TABLE I  
Fabric Constructions

<u>Sample No.</u>	<u>Polymer Inner Layer</u>	<u>Results</u>
1	PTFE*	Too thick - delaminated badly
2	90 poly(1-butene)-10 PTFE	Satisfactory
3	90 poly(ethylene-methyl acrylate)-10 PTFE	Shriveled during extraction
4	90 poly(ethylene vinyl acetate)-10 PTFE	Shriveled during extraction
5	90 polyethylene-10 PTFE	Good appearance, internally weak
6	90 polypropylene-10 PTFE	Too stiff

---

\* Poly(tetrafluoroethylene).

From the above test specimen preparations, eight samples were selected for submission to the Project Officer for designation of the preferred material for scale-up. The physical properties of the eight samples are shown in Table II.

Table II

## Laminated Fibrillar Materials

Sample No.	Composition*	Thickness, mils (avg.)	Weight oz/yd <sup>2</sup>	Hydrostatic Resistance**, psi.	Water Vapor Transmission***, g/24 hrs/m <sup>2</sup>
7-3	N-(PB + PTFE)-N	8.2	3.61	10-11 <sup>†</sup>	1130 <sup>†</sup>
7-4	N-(PB + PTFE)-N	7.7	3.45	10-11	1260
7-5	N-(PB + PTFE)-N	7.5	3.29	9-10	1270
8-2	D-(PB + PTFE)-D	9.0	3.92	7-8	670
9-2	N-(PE + PTFE)-N	8.4	3.79	11-12	650
12-1	N-(PP + PTFE)-N	8.9	3.60	7-8	600
13-3	D-(PP + PTFE)-D	13.0	5.12	8-9	460
13-4	D-(PP + PTFE)-D	13.3	5.01	3-4	620

\* PB is poly(1-butene), Mobil PB 001.

PE is polyethylene, Tenite 860A.

PP is polypropylene, Profax 6523.

PTFE is poly(tetrafluoroethylene), Teflon 30B dispersion.

N is nylon fabric, 1 oz/yd<sup>2</sup>.

D is dacron fabric, 1 oz/yd<sup>2</sup>.

\*\* Federal Test Method Std. 191-5512.

\*\*\* Federal Test Method Std. 406, Method 7032, Method B.

+ American Cyanamid Laboratories.

++ U. S. Army Natick Laboratories.

It is evident that the test data vary markedly between the two laboratories. At the time when the discrepancy emerged no explanation was readily apparent and the data of Natick were accepted as the correct values. It was later found that the difficulty was due to the use of a under-sized water reservoir. The distance between the test fabric and water surface was shorter than the 25 mm prescribed in the standard test procedure resulting in an increased rate of moisture vapor transmission.

On the basis of overall fabric properties including lamination integrity, flexibility, uniformity and wrinkle free appearance as well as the water control properties, polyester-90 poly(1-butene)-10 poly (tetrafluoroethylene)-polyester was designated for making the 100 yard run.

2. The approach to make OGL06 shade laminated fabric was via dyed fabric and pigmented polymer inner layer. Two hundred yards of nylon dyed OGL06 were purchased from Travis Fabrics of New York. Forty pounds of poly(1-butene) were pigmented by Customcolors, Inc., of Cumberland, Rhode Island. When these raw materials were converted into laminated fabric, the shade was lighter than desired. This was due to the fact that when poly(1-butene) was fibrillated during the process the high surface area resulted in light scattering and hence appeared lighter in this form than it did in the form of a thin continuous film. Improvement in color match needs to be sought by deepening the coloration. Two aspects of this effort were rewarding. Neither dye from the nylon nor pigment from the polymer were depleted during solvent extraction. Two 12 by 3 inch samples were supplied to NIABS.

Similar material was made from dyed polyester. Unfortunately, this dye was removed completely by the methylene chloride during solvent extraction. The obvious way to eliminate this difficulty is to specify the use of a dye unaffected by methylene chloride.

In addition to the routes to an OGL06 shade fabric described above, fabric specimens were submitted to the dyeing laboratory of Cyanamid. No standard dyeing procedure was found to simultaneously dye fabric and polymer inner layer. The most promising method depends on starting with dyed fabric and after the laminate has been produced to dye the polymer inner layer. In any event, more work remains to reach a satisfactory method of coloration.

#### C. Intermediate Scale-up

Concurrent with laboratory work on material optimization, effort proceeded to make ten yards 12 to 24 inch width nylon (1 oz/yd<sup>2</sup>)-fibrous 90 poly(1-butene)-10 polytetrafluoroethylene-nylon (1 oz/yd<sup>2</sup>). The intent of this task was to develop means to make the best material known into a sizable specimen. The prototype demonstration sample shown to the Natick Laboratories had been made with a laboratory press rather than by continuous extrusion-lamination.

Processing conditions were set based on laboratory experience but at a somewhat higher extrusion temperature to provide a margin of safety for the expensive plant machinery (See Appendix B). Laboratory tests showed that the fibrillating polymer-matrix-polymer feed stock would purge polyethylene from the extruder cleanly and further that upon completion of the run polyethylene would purge the composition so that it would not be necessary to disassemble the machinery before returning it to normal service. An external heater was brought to the facility (WRAPS, Inc., East Orange, New Jersey) and piped into the system to raise the roll temperature of the lamination station. Following these preparations the run was started and proceeded smoothly. Several settings of extruder screw speed, lamination roll speed and lamination pressure were tried. The preferred are listed in Table III.

The appearance of the laminated, but as yet unextracted product was smooth, uniform and acceptably thin (13 mils). No problem had been encountered in fabric feed or folding at the roll nip either when paper was used as a web or not. Lamination was more secure with paper support. However, as soon as the product was extracted to create the porosity essential for high moisture vapor transmission, it was clear the final laminated fabric was unsuitable. The inner layer was very weak and slight handling was sufficient to separate it into two layers. Hydrostatic resistance was essentially zero. The best 10 yards 24-inch wide section from the trial was submitted to the NIARS. Tests by the Natick Laboratories showed an average hydrostatic with resistance of 0.8 psi, moisture vapor transmission of 840 g/m<sup>2</sup>/24 hours, and a fabric weight of 4.1 oz/yd<sup>2</sup>.

Table III

Extrusion/Lamination Conditions

WRAPS, Inc. Run No. 1 (Specimen D)

Extrusion:

Temperature Profile on Machine;	
Flat, End-to-End,	430°F. (220°C.)
Melt Temperature at Die	443°F. (228°C.)
Screw Speed	25 rpm
Pressure (Generated by Valve at Outlet)	1800 psi
Output Rate	95 lbs./hr.

Lamination:

Roll Speed	7.5 fpm
Circulating Water Temperature	150°F. (66°C.)
Pneumatic Pressure on Rolls	35 psi (max = 100)

#### D. Process Development and Material Production

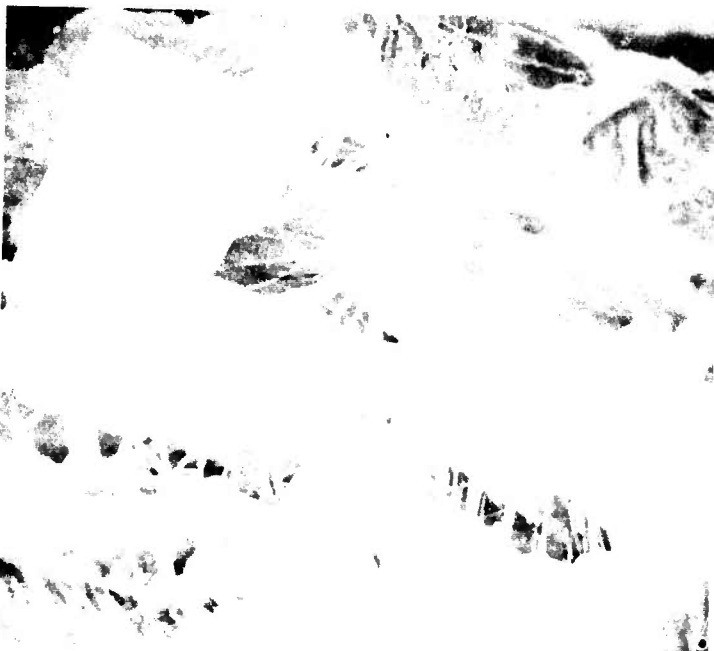
The third objective was to develop a process for continuous extrusion-lamination of the selected fabric-fibrous polymer-fabric material selected by the NIABS from Section B above, and to produce 100 yards of 36-inch minimum width of the laminate. Polyester (1 oz/ yd<sup>2</sup>)-90 poly(1-butene)10 poly(tetrafluoroethylene)-polyester (1 oz/yd<sup>2</sup>) was designated as the material of choice.

Experience of the Intermediate Scale Run described in Section C showed that mechanical processing including extrusion, cloth feed, and lamination was feasible. It was also found that product quality was badly deficient. The decision was to pursue development with the same subcontractor. Further to proceed to identify the processing conditions responsible for product failure.

Scanning electron micrographs of the laboratory laminated specimens and the WRAPS product showed scant fibrillar structure in the latter but "normal" fibrillarity in other samples. These are shown in Figures 1a and 1b. Poor fibrillar character would explain the low quality. Hence several processing and material variables were examined on laboratory equipment to identify causative factors including:

Extruder die gap	Pigmented feed stock vs. standard
Extruder worm speed	feed stock
Take away speed	Fibrillar loading
Lamination roll temperature	Matrix composition
Lamination roll pressure	Extruder temperature profile
	Cloth used for lamination

The most important factors were found to be type of cloth, temperature profile and extruder worm speed. The pigmented feed stock was somewhat inferior to the normal standard. On inquiry to Customcolors Inc. it was found that a dispersing aid had been used, so it was immediately decided to use unpigmented feed stock on the next large scale run. The cloth purchased for this run (heat set polyester) appeared to be the same as that used during the early, and successful, laboratory laminations hence would not constitute a problem. A face roll temperature of 90°C was found to eliminate weak lamination at standard roll pressure. A die gap of either 5 or 12 mils was found satisfactory. A poly(1-butene) loading of 35% was equal to the normal 27% loading. Since the WRAPS Run 1 product was more bulky and less dense than laboratory prepared material the PB loading was increased to 30%. The laboratory specimen made under preferred conditions withstood 20 psi liquid water (9 mils thickness) and had a wrinklefree surface.



Processed in Brabender at Stamford

Cross Section 1000X

a



Processed at WRAPS

Cross Section 1000X

b

Figure 1. Scanning Electron Photomicrographs of Pigmented Feed Polymer Compounded at Werner and Pfleiderer for Extrusion at WRAPS, Inc.

After all the information had been considered, the following plans for WRAPS Run 2 were adopted as a means of determining the way to make an optimum run of 100 yards.

1. Compound 150 pounds of PMMA-WSR 205 matrix,\* 30% fibrillar loading.
2. Compound 150 pounds of PMMA-WSR 301 matrix,\* 30% fibrillar loading.
3. Request a minimum die gap setting (15 mils promised).
4. Use an ascending temperature profile 170-200°C on the extruder.
5. Raise the steel roll temperature to maximum obtainable (~ 90°C expected).
6. Use pigmented feed stock on hand to purge polyethylene and line out the operation.
7. Switch over to feed stock No. 1 above.
8. Prepare fabric at several extruder worm speeds. (The valve between the extruder and the adapter open and the high machine speed to maximize shear while minimizing residence time.)
9. Seek the minimum thickness consonant with matrix-integrity.

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\* Poly(methyl methacrylate)-poly(ethylene oxide).

In practice it was not possible to follow all the elements of the plan. The die gap was 20 mils rather than 15. The extrudate could not be stretched far enough to make an 11 mil (estimated from the paper faced unextracted laminate) sheet. Unlike the previous run the extruded sheet was not uniform from one end of the die to the other. The center 40% appeared heterogeneous and lumpy. Invariably it was the first section to tear apart during increased stretching.

The cloth fed proceeded without difficulty. Lamination was secure even to the extent that the paper web was also fully bonded. Table IV summarizes the operating conditions used.

TABLE IV  
Operating Conditions of WRAPS Run 2

<u>Sample</u>	<u>Screw RPM/ Amps</u>	<u>Line FPM</u>	<u>Pneumatic Loading PSI</u>	<u>Paper</u>	<u>Miscellaneous</u>	<u>Amount (Est.) Ft.</u>	<u>Total* Thickness Mils</u>
A	125/76	19	15	Yes	(Untrimmed)	190	24-28
B	125/76	19	15	Yes	(Trimmed)	57	25-30
C	24/54	2.25	15	Yes	(Trimmed)	20	27-32
D	24/54	2.25	15	No	(Trimmed) (Lean streaks near center)	20	21-24
E	24/54	2.25	45	No	(Trimmed) (One edge <u>very</u> thick > 30 mils)	5	20-27

\* Two layers of paper are 9 mils thick.

All specimens made at a temperature profile with controllers at 390°F (199°C) except die and combining adapter at 425°F (219°C).



Small sections of the samples were extracted in the laboratory. Their physical properties appear in Table V.

TABLE V

Physical Properties of Laminated Fabric from WRAPS Run 2\*

<u>Sample No.</u>	<u>Weight oz./yd<sup>2</sup></u>	<u>Hydrostatic Water Resistance, psi</u>	<u>Water Vapor Permeability grs/m<sup>2</sup>/24 hrs.</u>
A1	6.5	14.0	550
A2	6.2	16.4	575

\* Determinations made by Natick Laboratories.

In summarizing the results of Run 2 at WRAPS, Inc., several main features should be noted. The mechanical processing once again proceeded smoothly. Product quality in terms of water control and in fabric integrity was greatly improved over Run 1. Cloth-polymer-cloth lamination was secure regardless of the operating conditions. The real difficulties were (1) inability to make integral fabric less than ~ 15 mils; and (2) a continuing partial degradation of coherent fibrillar structure in the inner polymer layer.

Laboratory experiments were directed toward the solution of the first problem. It was found that increasing the extruder screw speed, reduction in die gap and, to a lesser degree, an increase in polymer matrix temperature as it exited the die, were each beneficial in reaching a ten mil thickness. High extruder temperature and extended residence time in the extruder caused a reduction in fibrillar structure and hence hydrostatic water resistance. Personnel at WRAPS, Inc., were presented with two potential methods to obtain a narrower die gap (12 mils). One by determining the cost of simply tightening down the die with the knowledge that the edge seals would have to be replaced at a pre-determined cost, and secondly by tapering the die gap from the normal 20 mils at each end to 12 mils in the middle 36-inch section. It was agreed to follow the tapering method for the next run. To insure that no foreign particles would become lodged in the narrowed die the feasibility of incorporating a 40-60 mesh screen pack was demonstrated in the laboratory extruder.

For the selection of operating temperature it was now clear that a balance must be struck between retention of fibrillarity implying lower temperature and achieving thin extruded sheet which was known to be promoted by high temperature. Laboratory experiments demonstrated that processability as low as 150°C was possible without

excessive pressure build up and that with the extruder temperature at 170°C and a die temperature of 210°C thin stock could be produced. One variable which could not be assessed was the influence of the large volume adaptor between the extruder and the die in the WRAPS machinery.

With this information available it was proposed that one more attempt be made with commercial scale equipment. The necessary arrangements were made for raw materials and other services. The plan for Run 3 was as follows:

1. Obtain 50 pounds of poly(1-butene) pigmented at Customcolors, Inc., Cumberland, Rhode Island, at three times the pigment loading used in Run 1 but without dispersing aid.
2. Compound 150 pounds of pigmented polymer feed stock and 150 pounds unpigmented at Werner & Pfleiderer, Walldwick, New Jersey, both with 30% fibrillar loading in a PMMA-WSR301 matrix.
3. At the suggestion of the NLABS, use release paper as the carrying web during lamination.
4. Make sure the die gap is less than 15 mils.
5. Start the extrusion-lamination with pigmented feed stock and dyed polyester cloth.
6. Purge with unpigmented stock and repeat the process with white cloth in each case adjusting extruder screw speed and lamination speed to obtain the thinnest integral sheet possible.
7. Make at least 100 yards, 36-inch width with at least one 25 yard section at equilibrium conditions.

The operating conditions for Run 3 are listed in Table VI.

The principal operating observation made in Run 3 was that it was possible to make the thinnest material at slow extruder speed and slow take-away speed. This can be attributed to the rheological nature of the extruding polymer in that slower rate of strain results in maximum ultimate strain, i.e., stretchability. A die possessing a 3-inch parallel lip would therefore assist in permitting the production of thin laminate by forming the film and eliminating plastic memory.

At the completion of the extrusion-lamination the entire product was inspected. The sections selected for extraction are described in Table VII.

The most uniform sections were found among the unpigmented portions extracted. Physical data on three samples measured appear in Table VIII.

A total of 100 yards, 36-inch wide were extracted as described in Appendix B, Section IV, and delivered to NIABS.

The knowledge acquired during this project permits the fabrication, on a laboratory scale, of material superior to earlier products. Limited testing shows it to be  $\sim 3.2 \text{ oz/yd}^2$ , have hydrostatic resistance of 25 psi and water vapor permeability of  $880 \text{ grs/24 hrs/m}^2$ . Product made on a commercial scale does not equal these properties. Machine design explains the discrepancies.

E. Recommendations

It is recommended that future work be directed toward the following objectives:

1. Coloration of the fabric.
2. Increase of laminar strength.
3. Development of a commercial scale method for extraction.
4. Location or design and fabrication of extrusion equipment which permit duplication of laboratory process conditions.

Table VI

WRAPS Run No. 3

## Operating Conditions

Section	°F Machine Temperatures (Relative to Control Points)					Melt Temp. °F	Screw RPM	Amps	Extruder Pressure psi	Roll Speed fpm	Water Temp. °F	Pneum. Press. psi
	Barrel 1	Barrel 2	Barrel 3	Barrel 4	Barrel 5							
	1	2	3	4	5							
Feeds Tail	360	360	380	380	380	393	21.5	18	150	6-7	206	100
F <sub>21</sub>							21.5			6-7	---	100
E <sub>73</sub>							75			~	---	100
E <sub>50</sub>							50			..	---	100
D	340	340	355	355	355	361	25	34	160	5.5	208	100
A <sub>25</sub>	340	340	355	355	355	362	25	26	150	4.6	205	100

Roll surface 810C, (1730F)

Table VII

WRAPS Run No. 3

## Pre-Extraction White Products and Characteristics

Section	Length	Inside Edge	Thickness (Mils) Across Sheet; Start/Finish						Outside Edge	Remarks
F <sub>21.5</sub> Tail	27'	16-17	17.5/ 15.5	19.5/ 17.5	23/ 21	22.5/ 20.5	18/ 16	13.5/ 14	12-13	Profile High die T (435°F). Heavier portions intact (outer edge); Pinholes in lean portions. Gas bubbles (?) center section.
F <sub>21.5</sub>	28'	16-18	17.5/ 16	20/ 21	26/ 22.5	24/ 21.5	19.5/ 18	14.5/ 13	12.5-14	Profile High Die T. Some small gas bubbles like defects down center of sheet. Integrity is high except for lean outer edge (3") and bubbles near center.
E <sub>7.5</sub>	44'	16-18	17/ 16.5	15/ 18	20/ 21	16.5/ 19	17.5/ 17	15/ 14	12-14	Good integrity throughout, except occasional weakness (cheesey) in outer edge and occasional pinhole in inner half.
E <sub>30</sub>	32'	16-18	16/ 16.5	13/ 14.5	14.5/ 13.5	14/ 14	13/ 13	12/ 12	11.5-13.5	Inside half slightly cheesey, outside half clean, to holey edge. Occasional pinhole in outside half.
D	82'	16-18	17/ 17.5	14.5/ 14.5	15.5/ 14.5	14.5/ 14	14/ 14	12/ 12.5	11-12	Inside edge (3") heavy; outside edge (3") lean, occasional pinhole, porous; two 3-inch trims would yield a 25 yard 30 inch sheet, 14-16 mils thick of good integrity.

## Green Product

A <sub>25</sub>	75'	14-17	13.5/ 14	14/ 13.5	16/ 17	16/ 15.5	15/ 16	14.5/ 15	13-15	Always "cheesey" inner half; frequent pinholes in otherwise far superior outer half. Groups of holes show up sporadically or cyclically with relatively good but short (2'-3" section; in between.
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TABLE VIII

Physical Properties of Laminated Fabric from WRAPS Run 3

<u>Sample No.</u>	<u>Weight, oz/yd<sup>2</sup></u>	<u>Hydrostatic Water Resistance, psi</u>	<u>Water Vapor Permeability, grs/m<sup>2</sup>/24 hrs.</u>
E50	5.2	21.5	590
F21.5	4.8	11.5	704

## Summary

a. The functional and processing characteristics of six different fibrous, porous polymer structures were explored. The relative performance of nylon and polyester fabrics as top and bottom facing materials assessed. Eight of these samples of three-layer laminate construction were submitted to U. S. Army Natick Laboratories. NIABS designated poly(1-butene) 90%-tetrafluoroethylene 10% as the fibrous porous polymer structure and polyester as the top and bottom facing material for producing 100 yards, 36-inches wide.

b. A method was scaled up for continuous extrusion-lamination of an intermediate width (24 inches) of nylon (1 oz/yd<sup>2</sup>)-fibrous 90 poly(1-butene) 10 Teflon PTFE-nylon (1 oz/yd<sup>2</sup>). The linear yards of the material was produced for evaluation. The material from this first run on commercial equipment was of low quality showing almost no resistance to liquid water and little laminar integrity.

c. A process for continuous extrusion-lamination of the selected fabric-fibrous polymer-fabric material was developed, and 100 yards of 36-inch minimum width was produced on commercial scale equipment and delivered to the Government.

The mechanical operation proceeded without difficulty. A substantial portion of the product exhibited hydrostatic water resistance of 21.5 psi, moisture vapor transmission of 590 grams per square meter per 24 hours and total fabric weight of 5.2 ounces per square yard.

d. Material made in the laboratory extruder-laminator is superior to commercial scale production in terms of fabric integrity, weight, uniformity, and water control properties. This is believed attributable to the longer residence time necessitated by the equipment design and the extruder configuration of the commercial unit.

e. Efforts to produce finished fabric in OG106 shade were not successful. The best result was obtained in the laboratory with dyed nylon top and bottom facing material and pigmented fibrous polymer inner layer. A 25 yard piece of the 100 yard, 36-inch wide final product was also in OG106 shade. The dyed polyester used in its construction was not fast to the solvent used in the extraction.

f. The feasibility of large scale manufacture of a water vapor permeable, liquid water resistant material has been demonstrated. Additional equipment design and development effort will be required before adequate product quality control and production rate can be achieved.

g. A production technique must be developed for continuous extraction of the secondary resin.

## Appendix A

### Procedure for Laboratory Preparation of Laminated Fabric Test Specimens

#### Milling and Grinding

The compositions (see page 2 of text) were milled together on a Farrell 6-inch two roll mill heated by steam at 170°C. Typically 300 g of PMMA (Acrylite<sup>®</sup> H-12) was put on the hot rolls with the gap adjusted to less than the size of the granules and was allowed to stand 12-15 minutes before the rolls were started. Then as soon as the polymer had fluxed, 100 g of PEO (Polyox<sup>®</sup> WSR 205) was blended in in about 3 minutes and was followed a minute later by 135 g of polyolefin added in an equal time. Finally 15 g PTFE (25 g 60% Teflon<sup>®</sup> 30-B dispersion) was added in small portions in about 1 minute. During these additions, several adjustments of the roll spacing were required to maintain a roll of polymer blend in the nip of about 1/2 inch diameter.

As soon as the blend appeared reasonably uniform, it was removed from the rolls with the doctor blade, folded, and returned to the nip. This was done 6 times in about 5 minutes and was completed 30-45 minutes after the H-12 had been put on the rolls.

The blend was next removed from the rolls again and immediately cut into pieces roughly 2-inches square. After these had cooled to room temperature, they were passed through a small Cumberland grinder which broke the down to particles of 1/8-1/4 inch or less. The blend was bottled in this form and was oven dried (65°C) immediately before use.

#### Extrusion and Lamination

As shown schematically in Figure 1, the chopped and dried polymer blends were extruded in a 3/4 inch laboratory extruder (Brabender Model 250). The screw used had a uniform taper giving a 3:1 compression ratio and fed a 4 inch horizontal ribbon die with an adjustable gap. The extruded polymer was immediately laminated with top and bottom layers of fabric (1 oz/yd<sup>2</sup> nylon and dacron) by feeding the hot polymer and fabric directly into the top nip of a set of three 6 inch take-off rolls which were heated to a pre-determined temperature by hot water or steam. Pressure on the top nip was regulated by means of two 2 x 2 inch pneumatic cylinders with nitrogen pressures up to 90 psi, but the stops on the lower nip were adjusted to give a slight clearance between rolls and laminate. The roll speed was controlled by a Reves Moto-Drive unit which allowed continuous variation of roll surface speed from about 1.6 to about 10 feet/minute. After the laminate had passed over the middle roll, through the lower nip, and back around the bottom roll, it passed to a wind-up roll driven by a torque motor set to maintain moderate tension on the laminate.



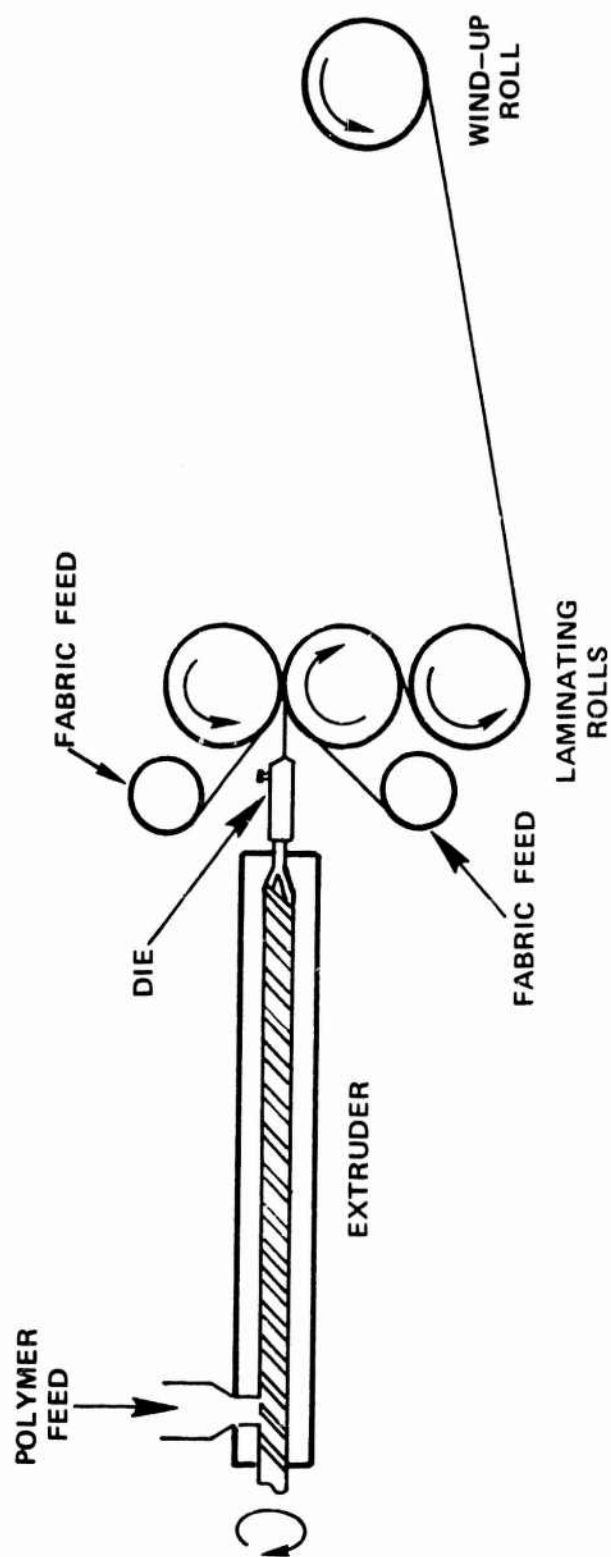


Figure A-1. Apparatus for Laboratory Extrusion and Lamination

### Extraction

Approximately 12 inch lengths of laminate were cut, measured for thickness, and marked with a 10 inch interval for subsequent shrinkage measurement. These specimens were next clipped with large paper clips to 10 inch squares of rigid perforated aluminum sheet which were then placed in a rack in a 5 gallon methylene chloride-filled extractor at room temperature. After standing about 3 hours with constant stirring of the solvent or overnight without stirring, the rack and samples were removed, allowed to drain briefly, and placed in a second extractor for an equal time. Specimens were then removed, separated from the aluminum sheets, and quickly hung up to air dry in the hood. During drying, the samples were kept under about 150 g tension to minimize shrinkage and wrinkling.

A maximum of 30 specimens was run through the first extractor before replacing the solvent. It was calculated that this would keep the dissolved polymer concentration below 1%, so that the residual polymer concentration in the second bath would be negligible for these samples. The times specified were not intended to be minimum times (1/2 hour in a stirred bath would probably be adequate), but were chosen for convenience and for certainty of achieving complete extraction.

## Appendix B

### Procedure for Large Scale Preparation of Laminated Sheet

#### I. Pre-blending

In order to insure uniform feed stock for the compounding operation, ingredients were pre-blended in 25-lb. lots in a 1 cu. ft. double ribbon blender. On extended runs separate feed streams to the compounding unit would be preferable.

#### II. Compounding

Using the Werner & Pfleiderer ZSK 53/2 at their Waldwick, N. J., facilities, pre-blend could be processed at about 80 lbs./hour when handling our most difficult formulation (containing WSR-301). Generally it was necessary to run the first two zones of the machine very hot to achieve early fluxing, followed by high shear mixing at reduced temperatures to achieve thorough dispersion and fibrillation.

Using the most intense mixing screw configuration available on this machine, and the following temperature profile, a uniform product was achieved:

#### Barrel Temp., °C

Zone 1	260
2	290
3	175
4	140
5	125
Die	145

Screw speed was held at maximum (300 rpm). Extruded strands were water-cooled and cut into pellets on a Cumberland cutter.

#### III. Extrusion/Lamination

A 3 1/2" Sterling extruder fitted with a 48" Flex Lip sheet die, delivering vertically downward, was used to convert the compounded stock to laminated sheet. This equipment is located at WRAPS, Inc., East Orange, N. J.

Compounded stock had to be pre-dried to less than 0.3% moisture to avoid decomposition of resins and gas/water vapor bubbles in the extruded sheet.

Using a 60 mesh screen pack, wide open back pressure valve, and the die gap as narrow as possible (12-13 mils), best results were achieved at a machine speed of 25 rpm, with the take up speed at 5.5 ft./min. (See Table of Operating Conditions, note Section "D".) Die swell at higher speeds obviated thin sheet production.

Square weave polyester (1.0 oz/yd<sup>2</sup>) was fed from tensioned unwind stations to each side of the extruded sheet at the nip of the hot (208°F) laminating roll which carried the maximum pneumatic loading (> 100 psi).

Stock was trimmed to about 36" and taken up on a turret winder.

#### IV. Extraction

A jelly-roll configuration was constructed of the unextracted laminate-sulfite paper - 1/4" mesh hardware cloth - sulfite paper. The consolidated roll was immersed in methylene chloride (two, 55 gallon drums welded into a cylinder 48" tall to serve as the extraction vessel) and soaked overnight. During the next 2 days the entire roll was raised from the solvent, allowed to drain, and reimmersed at intervals of 4 hours. It was then transferred to fresh solvent, re-equilibrated, rinsed in a third container of fresh solvent and air dried.